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(71) Applicant: Shin-Etsu Chemical Co., Ltd.
Chiyoda-ku, Tokyo (JP)

(72) Inventors:
• Nakamura, Hajime,
Magnetic Materials Rsrch. Center
Echizen-shi, Fukui-ken (JP)

- Hirota, Koichi,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)
- Shimao, Masanobu,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)
- Minowa, Takehisa,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)

(74) Representative: Stoner, Gerard Patrick et al
Mewburn Ellis LLP
York House
23 Kingsway
London WC2B 6HP (GB)

(54) Rare earth permanent magnet

(57) A rare earth permanent magnet is in the form of a sintered magnet body having a composition $R^1_a R^2_b T_c A_d F_e O_f M_g$ wherein F and R² are distributed such that their concentration increases on the average from the center toward the surface of the magnet body, the concentration of R²/(R¹+R²) contained in grain boundaries surrounding primary phase grains of (R¹, R²)₂T₁₄A tetragonal system within the sintered magnet body is on the average higher than the concentration of

R²/(R¹+R²) contained in the primary phase grains, and the oxyfluoride of (R¹, R²) is present at grain boundaries in a grain boundary region that extends from the magnet body surface to a depth of at least 20 μm. The invention provides R-Fe-B sintered magnets which exhibit high magnet performance despite minimal amounts of Tb and Dy used.

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$$H_{cj} \geq 1 + 0.2 \times M$$

wherein H_{cj} is a coercive force in unit MA/m and M is the content (wt%) of element M in the overall magnet and $0.05 \leq M \leq 10$. This method, however, is extremely unproductive and impractical.

[0009] An object of the present invention is to provide new and useful R-Fe-B permanent magnets (wherein R is at least two selected from rare earth elements inclusive of Sc and Y) which exhibit high performance despite small amounts of Tb and Dy used, and methods of making such magnets.

[0010] Regarding R-Fe-B sintered magnets (wherein R is one or more elements selected from rare earth elements inclusive of Sc and Y), typically Nd-Fe-B sintered magnets, the inventors have found that when a magnet body is heated at a temperature not higher than a sintering temperature, with a powder based on a fluoride of Dy and/or Tb packing the magnet body surface, both Dy and/or Tb and fluorine which have been in the powder are efficiently absorbed by the magnet body, and Dy and/or Tb is enriched only in proximity to interfaces between grains to enhance an anisotropic magnetic field only in proximity to interfaces, for thereby enhancing a coercive force while restraining diminution of remanence. This approach is successful in reducing the amount of Dy and Tb used as well.

[0011] Accordingly, the present invention provides a rare earth permanent magnet in the form of a sintered magnet body having an alloy composition $R^1_a R^2_b T_c A_d F_e O_f M_g$ wherein R^1 is at least one element selected from rare earth elements inclusive of Sc and Y and exclusive of Tb and Dy, R^2 is one or both of Tb and Dy, T is one or both of iron and cobalt, A is one or both of boron and carbon, F is fluorine, O is oxygen, and M is at least one element selected from the group consisting of Al, Cu, Zn, In, Si, P, S, Ti, V, Cr, Mn, Ni, Ga, Ge, Zr, Nb, Mo, Pd, Ag, Cd, Sn, Sb, Hf, Ta, and W, a through g indicative of atom percents of the corresponding elements in the alloy have values in the range: $10 \leq a+b \leq 15$, $3 \leq d \leq 15$, $0.01 \leq e \leq 4$, $0.04 \leq f \leq 4$, $0.01 \leq g \leq 11$, the balance being c , the magnet body having a center and a surface. Constituent elements F and R^2 are distributed such that their concentration increases on the average from the center toward the surface of the magnet body. Grain boundaries surround primary phase grains of $(R^1, R^2)_2 T_{14} A$ tetragonal system within the sintered magnet body. The concentration of $R^2/(R^1+R^2)$ contained in the grain boundaries is on the average higher than the concentration of $R^2/(R^1+R^2)$ contained in the primary phase grains. The oxyfluoride of (R^1, R^2) is present at grain boundaries in a grain boundary region that extends from the magnet body surface to a depth of at least 20 μm .

[0012] In a preferred embodiment, the oxyfluoride of (R^1, R^2) at grain boundaries contains Nd and/or Pr, and an atomic ratio of Nd and/or Pr to (R^1+R^2) contained in the oxyfluoride at grain boundaries is higher than an atomic ratio of Nd and/or Pr to (R^1+R^2) contained at grain boundaries excluding the oxyfluoride and the oxide of R^3 wherein R^3 is at least one element selected from rare earth elements inclusive of Sc and Y.

[0013] In preferred embodiments, R^1 comprises at least 10 atom% of Nd and/or Pr; T comprises at least 60 atom% of iron; and A comprises at least 80 atom% of boron.

[0014] The present invention is successful in providing R-Fe-B sintered magnets which exhibit high magnet performance despite minimal amounts of Tb and Dy used.

BRIEF DESCRIPTION OF THE DRAWINGS

[0015]

FIGS. 1a and 1b are photomicrographs showing a Tb distribution image of a magnet body M1 manufactured in Example 1 and a Tb distribution image of a magnet body P1 as machined and heat treated, respectively.

FIG. 2 is a graph in which the average concentrations of Tb (a) and F (b) in the magnet body M1 of Example 1 are plotted relative to a depth from the magnet surface.

FIG. 3a, 3b, and 3c are photomicrographs showing compositional distribution images of Nd, O, and F in the magnet body M1 of Example 1, respectively.

FURTHER EXPLANATIONS, OPTIONS AND PREFERENCES

[0016] The rare earth permanent magnet of the present invention is in the form of a sintered magnet body having an alloy composition of the formula (1).

- and W. M may be contained in an amount of 0.01 to 11 atom%, and preferably 0.1 to 5 atom% of the overall alloy. The balance is composed of incidental impurities such as N and O.

[0029] The mother alloy is typically prepared by melting metal or alloy feeds in vacuum or an inert gas atmosphere, typically argon atmosphere, and casting the melt into a flat mold or book mold or strip casting. A possible alternative is a so-called two-alloy process involving separately preparing an alloy approximate to the $R_2Fe_{14}B$ compound composition constituting the primary phase of the relevant alloy and an R-rich alloy serving as a liquid phase aid at the sintering temperature, crushing, then weighing and mixing them. Notably, the alloy approximate to the primary phase composition is subjected to homogenizing treatment, if necessary, for the purpose of increasing the amount of the $R_2Fe_{14}B$ compound phase, since α -Fe is likely to be left depending on the cooling rate during casting and the alloy composition. The homogenizing treatment is a heat treatment at 700 to 1,200°C for at least one hour in vacuum or in an Ar atmosphere. To the R-rich alloy serving as a liquid phase aid, a so-called melt quenching or strip casting technique is applicable as well as the above-described casting technique.

[0030] The mother alloy is generally crushed to a size of 0.05 to 3 mm, preferably 0.05 to 1.5 mm. The crushing step uses a Brown mill or hydriding pulverization, with the hydriding pulverization being preferred for those alloys as strip cast. The coarse powder is then finely divided to a size of generally 0.2 to 30 μ m, preferably 0.5 to 20 μ m, for example, by a jet mill using nitrogen under pressure. The oxygen content of the sintered body can be controlled by admixing a minor amount of oxygen with the pressurized nitrogen at this point. The oxygen content of the final sintered body, which is given as the oxygen introduced during the preparation of the ingot plus the oxygen taken up during transition from the fine powder to the sintered body, is preferably 0.04 to 4 atom%, more preferably 0.04 to 3.5 atom%.

[0031] The fine powder is then compacted under a magnetic field on a compression molding machine and placed in a sintering furnace. Sintering is effected in vacuum or in an inert gas atmosphere usually at a temperature of 900 to 1,250°C, preferably 1,000 to 1,100°C. The thus sintered magnet contains 60 to 99 vol%, preferably 80 to 98 vol% of the tetragonal $R_2Fe_{14}B$ compound as a primary phase, the balance being 0.5 to 20 vol% of an R-rich phase, 0 to 10 vol% of a B-rich phase, 0.1 to 10 vol% of R oxide, and at least one of carbides, nitrides and hydroxides of incidental impurities or a mixture or composite thereof.

[0032] The sintered magnet body (or sintered block) is machined to a predetermined shape, after which a powder containing the fluoride of Tb and/or Dy is disposed on the surface of the magnet body. The magnet body packed with the fluoride powder is heat treated in vacuum or in an atmosphere of inert gas such as Ar or He at a temperature of not higher than the sintering temperature (referred to as T_s), especially 200°C to $(T_s-5)^\circ\text{C}$ for about 0.5 to 100 hours. Through the heat treatment, the fluoride of Tb and/or Dy is infiltrated in the magnet and the rare earth oxide within the sintered magnet body reacts with fluorine to make a chemical change into an oxyfluoride. The amount of fluorine absorbed in the magnet body at this point varies with the composition and particle size of the powder used, the proportion of the powder occupying the magnet surface-surrounding space during the heat treatment, the specific surface area of the magnet, the temperature and time of the heat treatment although the absorbed fluorine amount is preferably 0.01 to 4 atom%, more preferably 0.05 to 3.5 atom%. At this point, the absorbed Tb and/or Dy component concentrates adjacent to the grain boundaries.

[0033] The powder fed to the surface of the sintered magnet body may consist solely of the fluoride of Tb and/or Dy although in practice the magnet can still be manufactured as long as the powder contains at least 15% by weight, especially at least 30% by weight of the fluoride of Tb and/or Dy. Suitable components of the powder other than the fluoride of Tb and/or Dy include fluorides of other rare earth elements such as Nd and Pr, oxides, oxyfluorides, carbides, hydrides, hydroxides, oxycarbides, and nitrides of rare earth elements inclusive of Tb and Dy, fine powders of boron, boron nitride, silicon, carbon or the like, and organic compounds such as stearic acid.

[0034] The amount of the powder fed to the surface of the sintered magnet body may be about 0.1 to about 100 mg/cm^2 , preferably about 0.5 to about 50 mg/cm^2 of the surface.

[0035] Preferably the magnet body is further subjected to aging treatment.

[0036] The oxyfluoride of R (rare earth elements inclusive of Sc and Y) within the magnet is typically ROF, although it generally denotes oxyfluorides containing R, oxygen and fluorine that can achieve the effect of the invention including RO_mF_n (wherein m and n are positive numbers) and modified or stabilized forms of RO_mF_n wherein part of R is replaced by a metal element.

[0037] The thus obtained permanent magnet material containing the oxyfluoride of R can be used as a high-performance permanent magnet.

[0038] Methods of making such magnets, as disclosed herein, are an aspect of our invention.

EXAMPLE

[0039] Examples of the present invention are given below by way of illustration and not by way of limitation.

30 seconds while sonicating the suspension. The dysprosium fluoride powder had an average particle size of 10 μm . The magnet was taken up and placed in a vacuum desiccator where it was dried at room temperature for 30 minutes while evacuating by a rotary pump.

[0050] The magnet body packed with dysprosium fluoride was subjected to heat treatment in an Ar atmosphere at 800°C for 10 hours and then aging treatment at 510°C for one hour, and quenched, obtaining a magnet body within the scope of the invention. This magnet body is designated M2. For comparison purposes, a magnet body was prepared by effecting heat treatment without the dysprosium fluoride package. This is designated P2.

[0051] The magnet bodies M2 and P2 were measured for magnetic properties (Br, H_{cj}, (BH)_{max}), with the results also shown in Table 1. The compositions of the magnets are shown in Table 2. The magnet M2 of the invention marked a coercive force increase of 520 kAm⁻¹ relative to the coercive force of the magnet P2 having undergone heat treatment without the dysprosium fluoride package while showing a remanence decline of 5 mT. The distributions of Dy and F in the magnet M2 as analyzed by EPMA were equivalent to the distributions of Tb and F in Example 1.

Example 3 and Comparative Example 3

[0052] An alloy in thin plate form consisting of 12.5 atom% Nd, 1.5 atom% Dy, 0.5 atom% Al, 5.8 atom% B, and the balance of Fe was prepared by using Nd, Dy, Al, and Fe metals of at least 99 wt% purity and ferroboration, high-frequency melting them in an Ar atmosphere, and casting the melt onto a single chill roll of copper (strip casting technique). The alloy was exposed to hydrogen under 0.11 MPa at room temperature for hydriding, heated up to 500°C for partial dehydriding while evacuating the chamber to vacuum, cooled down, and sieved, obtaining a coarse powder of under 50 mesh.

[0053] On a jet mill using nitrogen gas under pressure, the coarse powder was finely divided into a powder with a mass base median diameter of 4.0 μm . The fine powder was oriented in a magnetic field of 15 kOe under a nitrogen atmosphere and compacted under a pressure of about 1 ton/cm². The compact was then placed in a sintering furnace with an Ar atmosphere where it was sintered at 1,060°C for 2 hours, obtaining a magnet block. Using a diamond cutter, the magnet block was machined on all the surfaces to dimensions of 10 mm x 10 mm x 3 mm thick. The magnet body was successively washed with alkaline solution, deionized water, nitric acid and deionized water, and dried.

[0054] Subsequently the magnet body was immersed in a suspension of 50 wt% terbium fluoride in ethanol for 30 seconds while sonicating the suspension. The terbium fluoride powder had an average particle size of 5 μm . The magnet was taken up and immediately dried with hot air blow.

[0055] The magnet body packed with terbium fluoride was subjected to heat treatment in an Ar atmosphere at 800°C for 10 hours and then aging treatment at 585°C for one hour, and quenched, obtaining a magnet body within the scope of the invention. This magnet body is designated M3. For comparison purposes, a magnet body was prepared by effecting heat treatment without the terbium fluoride package. This is designated P3.

[0056] The magnet bodies M3 and P3 were measured for magnetic properties (Br, H_{cj}, (BH)_{max}), with the results also shown in Table 1. The compositions of the magnets are shown in Table 2. The magnet M3 of the invention marked a coercive force increase of 750 kAm⁻¹ relative to the coercive force of the magnet P3 having undergone heat treatment without the terbium fluoride package while showing a remanence decline of 5 mT. The distributions of Tb and F in the magnet M3 as analyzed by EPMA were equivalent to those in Example 1.

Examples 4-8 and Comparative Examples 4-8

[0057] An alloy in thin plate form consisting of 11.5 atom% Nd, 2.0 atom% Pr, 0.5 atom% Al, 0.3 atom% Cu, 0.5 atom% M' (= Cr, V, Nb, Ga or W), 5.8 atom% B, and the balance of Fe was prepared by using Nd, Pr, Al, Fe, Cu, Cr, V, Nb, Ga, and W metals of at least 99 wt% purity and ferroboration, high-frequency melting them in an Ar atmosphere, and casting the melt onto a single chill roll of copper (strip casting technique). The alloy was exposed to hydrogen under 0.11 MPa at room temperature for hydriding, heated up to 500°C for partial dehydriding while evacuating the chamber to vacuum, cooled down, and sieved, obtaining a coarse powder of under 50 mesh.

[0058] On a jet mill using nitrogen gas under pressure, the coarse powder was finely divided into a powder with a mass base median diameter of 4.7 μm . The fine powder was oriented in a magnetic field of 15 kOe under a nitrogen atmosphere and compacted under a pressure of about 1 ton/cm². The compact was then placed in a sintering furnace with an Ar atmosphere where it was sintered at 1,060°C for 2 hours, obtaining a magnet block. Using a diamond cutter, the magnet block was machined on all the surfaces to dimensions of 5 mm x 5 mm x 2.5 mm thick. The magnet body was successively washed with alkaline solution, deionized water, citric acid and deionized water, and dried.

[0059] Subsequently the magnet body was immersed in a suspension of 50 wt% a 50:50 (weight ratio) dysprosium fluoride/dysprosium oxide mix in ethanol for 30 seconds while sonicating the suspension. The dysprosium fluoride and dysprosium oxide powders had an average particle size of 5 μm and 1 μm , respectively. The magnet was taken up and placed in a vacuum desiccator where it was dried at room temperature for 30 minutes while evacuating by a rotary pump.

(continued)

			Pr [at. %]	Nd [at. %]	Tb [at. %]	Dy [at. %]	Fe+Co [at. %]	B [at. %]	F [at. %]	O [at. %]	M* [at. %]
5	Example 8	M8	1.951	11.220	0.000	0.114	78.611	5.745	0.336	0.734	1.288
	Comparative Example 1	P1	1.958	11.259	0.000	0.000	79.412	5.765	0.000	0.810	0.795
10	Comparative Example 2	P2	0.000	13.883	0.394	0.000	77.956	5.797	0.000	0.623	0.747
	Comparative Example 3	P3	0.000	12.298	0.000	1.495	79.586	5.793	0.000	0.328	0.499
15	Comparative Example 4	P4	1.957	11.253	0.000	0.000	78.847	5.762	0.000	0.890	1.291
	Comparative Example 5	P5	1.960	11.271	0.000	0.000	78.977	5.771	0.000	0.727	1.294
20	Comparative Example 6	P6	1.955	11.244	0.000	0.000	78.783	5.757	0.000	0.970	1.290
	Comparative Example 7	P7	1.962	11.280	0.000	0.000	79.041	5.776	0.000	0.646	1.295
25	Comparative Example 8	P8	1.960	11.270	0.000	0.000	78.966	5.770	0.000	0.740	1.293
* Total amount of element as M in formula (1).											

[0063] Analytical values of rare earth elements were determined by entirely dissolving samples (prepared as in Examples and Comparative Examples) in aqua regia, and effecting measurement by inductively coupled plasma (ICP), analytical values of oxygen determined by inert gas fusion/infrared absorption spectroscopy, and analytical values of fluorine determined by steam distillation/Alfusone colorimetry.

Claims

1. A rare earth permanent magnet in the form of a sintered magnet body having an alloy composition $R^1_a R^2_b T_c A_d F_e O_f M_g$ wherein R^1 is at least one element selected from rare earth elements, Sc and Y, but not including Tb or Dy, R^2 is one or both of Tb and Dy, T is one or both of iron and cobalt, A is one or both of boron and carbon, F is fluorine, O is oxygen, and M is at least one element selected from Al, Cu, Zn, In, Si, P, S, Ti, V, Cr, Mn, Ni, Ga, Ge, Zr, Nb, Mo, Pd, Ag, Cd, Sn, Sb, Hf, Ta, and W, and indices a to g, indicating atom percents of the corresponding elements in the alloy, have values satisfying: $10 \leq a+b \leq 15$, $3 \leq d \leq 15$, $0.01 \leq e \leq 4$, $0.04 \leq f \leq 4$, $0.01 \leq g \leq 11$, the balance being c, said magnet body having a center and a surface, wherein constituent elements F and R^2 are distributed such that their concentration increases on the average from the center toward the surface of the magnet body, grain boundaries surround primary phase grains of $(R^1, R^2)_2 T_{14} A$ tetragonal system within the sintered magnet body, the R^2 concentration $R^2/(R^1+R^2)$ contained in the grain boundaries is on the average higher than the R^2 concentration $R^2/(R^1+R^2)$ contained in the primary phase grains, and the oxyfluoride of (R^1, R^2) is present at grain boundaries in a grain boundary region that extends from the magnet body surface to a depth of at least 20 μm .
2. The rare earth permanent magnet of claim 1 wherein the oxyfluoride of (R^1, R^2) at grain boundaries contains Nd and/or Pr, and an atomic ratio of Nd and/or Pr to (R^1+R^2) contained in the oxyfluoride at grain boundaries is higher than an atomic ratio of Nd and/or Pr to (R^1+R^2) contained at grain boundaries excluding the oxyfluoride and the oxide of R^3 wherein R^3 is at least one element selected from rare earth elements inclusive of Sc and Y.
3. The rare earth permanent magnet of claim 1 or 2 wherein R^1 comprises at least 10 atom% of Nd and/or Pr.

FIG.1

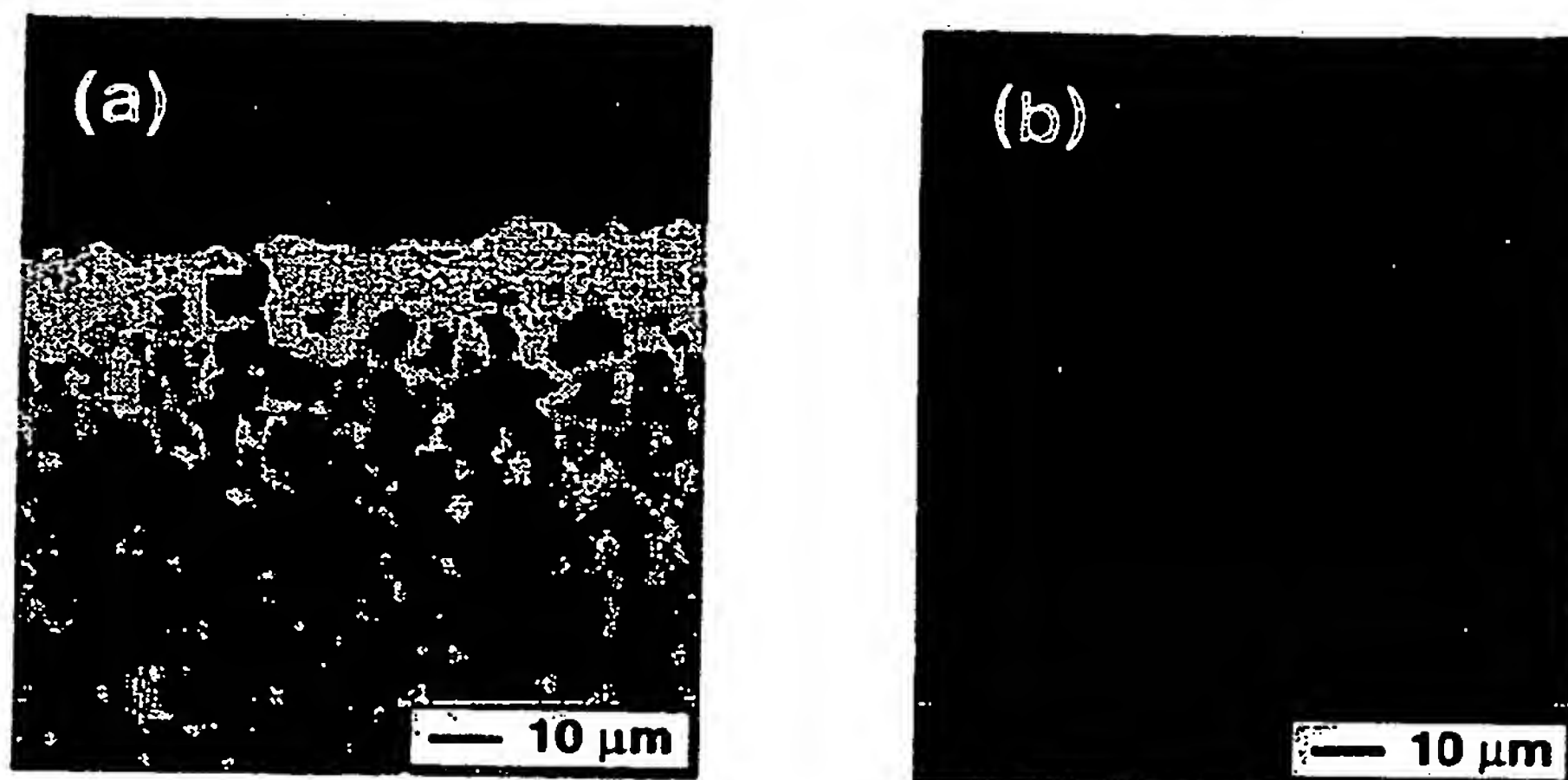
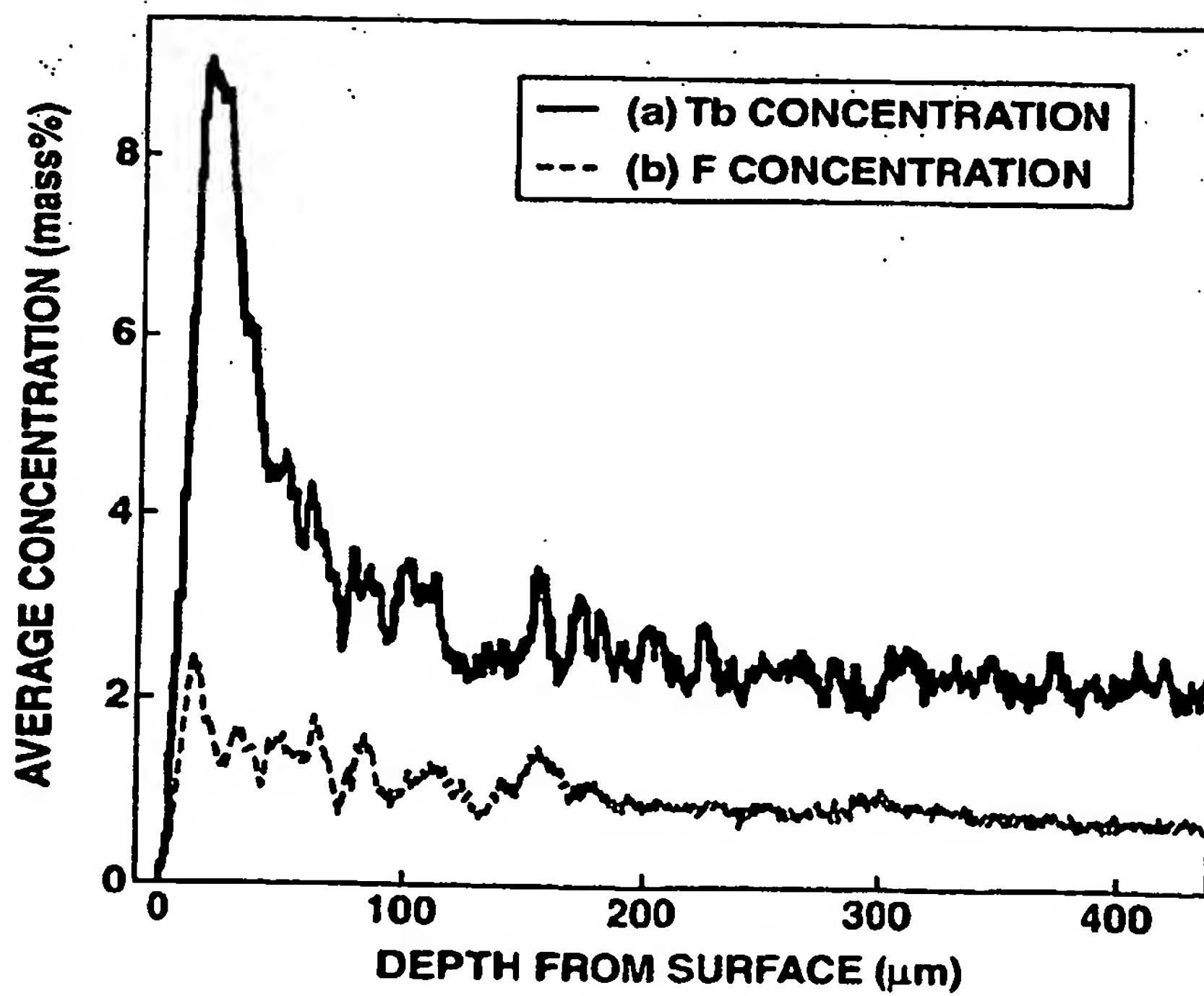


FIG.2



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(71) Applicant: Shin-Etsu Chemical Co., Ltd.
Chiyoda-ku, Tokyo (JP)

(72) Inventors:
• Nakamura, Hajime,
Magnetic Materials Rsrch. Center
Echizen-shi, Fukui-ken (JP)

- Hirota, Koichi,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)
- Shlmao, Masanobu,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)
- Minowa, Takehisa,
Magnetic Materials Rsrch. Center
Echizen-shi,
Fukui-ken (JP)

(74) Representative: Stoner, Gerard Patrick et al
Mewburn Ellis LLP
York House
23 Kingsway
London WC2B 6HP (GB)

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$R^2/(R^1+R^2)$ contained in the primary phase grains, and the oxyfluoride of (R^1, R^2) is present at grain boundaries in a grain boundary region that extends from the magnet body surface to a depth of at least 20 μm . The invention provides R-Fe-B sintered magnets which exhibit high magnet performance despite minimal amounts of Tb and Dy used.

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**ANNEX TO THE EUROPEAN SEARCH REPORT
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